Point-contact study of the $LuNi_2B_2C$ borocarbide superconducting film

O. E. Kvitnitskaya, Yu. G. Naidyuk, I. K. Yanson
B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences of Ukraine, 47 Lenin Ave., 61103, Kharkiv, Ukraine

T. Niemeier, G. Fuchs, B. Holzapfel and L. Schultz
Leibniz-Institut für Festkörper- und Werkstofforschung Dresden e.V., Postfach 270116, D-01171 Dresden, Germany

(Dated: October 1, 2010)

We present point-contact (PC) Andreev-reflection measurements of a superconducting epitaxial *c*-axis oriented nickel borocarbide film $LuNi_2B_2C$ (T_c=15.9 K). The averaged value of the superconducting gap is found to be $\Delta \simeq 2.6\pm0.2$ meV in the one-gap approach, whereas the two-gap approach results in $\Delta_1 \simeq 2.14\pm0.36$ meV and $\Delta_2 \simeq 3\pm0.27$ meV. The better fit of the Andreev-reflection spectra for the $LuNi_2B_2C$–Cu PC obtained by the two-gap approach provides evidence for multiband superconductivity in $LuNi_2B_2C$. For the first time, PC electron-phonon interaction (EPI) spectra have been measured for this compound. They demonstrate pronounced phonon maximum at 8.5±0.4 meV and a second shallow one at 15.8±0.6 meV. The electron-phonon coupling constant λ estimated from the PC EPI spectra turned out to be small (with $\lambda_{PC} \sim 0.1$), like in other superconducting rare-earth nickel borocarbides. Possible reasons for this are discussed.

PACS numbers: 72.10.Di, 74.45.+c, 74.70Dd

INTRODUCTION

Among the rare-earth nickel borocarbide superconducting $ReNi_2B_2C$ family, the system with Re = Lu belongs to the nonmagnetic ones with the highest T_c of about 16 K. Although numerous experiments were undertaken to study the superconducting state there, still there is space for more detailed investigation. This is most probably due to the complicated band structure in this compound what may certainly lead to the anisotropy of the superconducting gap or/and multi-band (gap) superconductivity. Point-contact (PC) Andreev reflection spectroscopy is a direct tool to clarify the superconducting gap characteristics, i.e. its value and its anisotropy as well as the temperature and magnetic field dependencies of the gap. Furthermore, PC spectroscopy itself provides a straightforward information as to the PC electron-phonon interaction (EPI) function $\alpha^2F(\omega)$ [1], which can be a test for the phonon-mediated superconductivity.

Several PC studies were performed on single crystals of $LuNi_2B_2C$ [2, 3]. These experiments have shown an anisotropy of the superconducting order parameter (gap). In Ref. [2], the authors claimed that the data are in favor of a two-gap model. A similar conclusion was made also by a study of single crystals and films of the related compound YNi_2B_2C in [4–6]. In this study, we present PC spectroscopy data for epitaxial c-axis oriented $LuNi_2B_2C$ films in order to compare these data with the results obtained for $LuNi_2B_2C$ single crystals and to receive information about EPI in $LuNi_2B_2C$.

EXPERIMENTAL DETAILS

High-quality $LuNi_2B_2C$ epitaxial c-axis oriented films with T_c = 15.9 K have been fabricated using pulsed laser deposition. The details of the film preparation are described in [7]. XRD measurements show almost perfect c-axis texture. The films show good homogeneity, very high in-plane and out-of-plane order and high enough for the thin films residual resistivity ratios up to 17 at film thicknesses of 250 nm. The quality of these films compared to single crystals was tested using a sample in the as-grown state for B_{c2} measurements. Upper critical field values in the <001> crystallographic direction are higher than those in single crystals ($B_{c2} \simeq 8$T [8]) and show a less pronounced S-shape behavior.

The PCs were established along the c-direction by the standard "needle-anvil" method [1] touching of the film surface by a sharpened Cu wire. The differential resistance $dV/dI(V)$ and the second derivative $d^2V/dI^2(V)$ were recorded by sweeping the dc current I on which a small ac current i was superimposed using the standard lock-in technique. The measurements were performing in the temperature range T=1.5-20 K and in magnetic fields up to 9 T.

PCS of superconducting energy gap

Spectroscopic information about superconducting energy gap is available in the case if the contact diameter d is smaller than the inelastic electron mean-free path as well as than the coherence length $\xi(0)$. $LuNi_2B_2C$ is characterized at low temperatures by coherence length about $\xi \approx 6$ nm [8] and elastic electron mean-free path $l \simeq 10$ nm evaluated from the mentioned below ρl value.

Estimation of the contact size from its resistance $R_{\rm PC}$ using the Wexler formula [1]

$$R_{\rm PC} \simeq (16\rho l)/3\pi d^2) + \rho/2d, \qquad (1)$$

gives $d \leq 10\,$nm for the typical PC resistance $R_{\rm PC}$=10 Ω with $\rho \simeq 2.7\mu\Omega$cm and $\rho l \simeq 3.6 \cdot 10^{12}\Omega \cdot$ cm^2 [9]. Thus, all the mentioned lengths are of the same order of magnitude, therefore it is not possible to say a priori whether investigated PCs are in the ballistic (or diffusive), in other words, in the spectroscopic regime. Independent of the PC resistance (of course the higher the resistance the higher probability to be in spectroscopic regime), each PC spectra should be tested in order to display the spectroscopic features both in the normal (phonons) and superconducting (gap minima) state.

We were able to obtain PC dV/dI characteristics, which demonstrate clear Andreev-reflection (gap) structures – pronounced minima at $V \simeq \pm\Delta$ at $T <$T$_c$ as it is shown in Fig. 1. For the investigated PCs the temperature of the vanishing of the superconducting main minimum in dV/dI was close to 16 K, that is, close to T$_c$ of the $LuNi_2B_2C$ film, testifying that superconductivity in the PCs is not degraded. To retrieve the superconducting gap value Δ and other parameters from the Andreev-reflection spectra the generalized Blonder-Tinkham-Klapwijk (BTK) theory [10] is commonly used. The application of the one-gap BTK model to the dV/dI curves from Fig. 1(bottom panel) results in a moderate fit to the experimental curves. Furthermore, the parameter Γ, which implies a finite lifetime of carriers due to inelastic scattering of charge carriers was found to be rather high (about half of the Δ value) for this PC. Such high Γ as compared to Δ can be connected with the anisotropy of the superconducting gap or a possible two-gap superconductivity. The two-gap(band) model is supported by a recent three-dimensional study of the Fermi surface of $LuNi_2B_2C$ [15], where contribution to the density-of-states (DoS) at the Fermi energy from 3 bands equal 0.24%, 22.64% and 77.1%, respectively, was found. That is, two bands basically contribute to DoS.

As we can see from Fig. 1(bottom panel) the fitting within the two-gap model gives a much better agreement with the experimental dV/dI data. Applying the two-gap fit the temperature dependencies of the superconducting gaps Δ, the smearing parameter Γ and the barrier strength Z were established (see Fig. 2). The so-called scaling parameter S corresponding to the ratio of the experimental dV/dI intensity to the calculated one reflects the quality of the fit and was tried to keep constant and close to 1 at fitting dV/dI. It turned out that the contribution K (or weight factor) of both gaps to the spectra is nearly equal K=0.5 and the fitting is possible with Γ=0. The latter gives strong support for the used two-gap model which confirms the multiband superconducting state in this compound. We should also note, that with increasing temperature the double minimum structure of dV/dI smears out above 11 K and this gives more space for the fitting parameters. Therefore their values are less precise by approaching T$_c$.

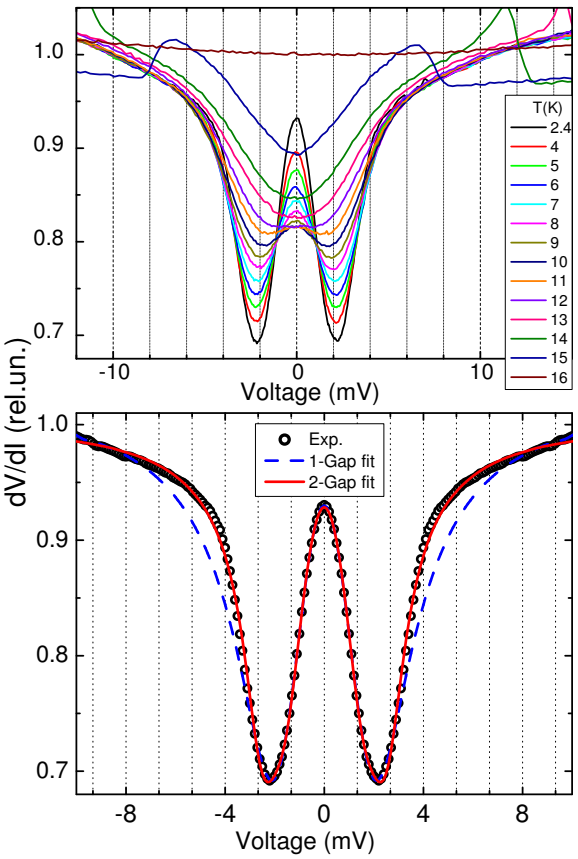

FIG. 1: Upper panel: dV/dI curves of a $LuNi_2B_2C$ - Cu contact ($R = 6.3\,\Omega$) established along the c-axis for varying temperature. Bottom panel: symmetrized and normalized to the normal state dV/dI curve at $T = 2.4\,$K (points) together with calculated curves according to the generalized BTK theory: dashed and solid lines - one-gap and two-gap fits, respectively.

The data for the two superconducting gaps presented in Fig. 2 well agree with results reported for $LuNi_2B_2C$ single crystals for the c-direction [2]. The averaged gap value of $\Delta \simeq 2.6\pm0.2\,$meV for all contacts (about 30) calculated in the one-band approach turned out to be close to the averaged gap of $\Delta \simeq 2.4$–$2.5\,$meV reported for PCs on single crystals for the c-direction [2, 3].Thus, the quality of the investigated films is comparable to that of the best single crystals. The mean gap values $\Delta_1 \simeq 2.14\pm0.36\,$meV and $\Delta_2 \simeq 3\pm0.27\,$meV established for about 15 PCs from the two-gap approach are also in line with data obtained for $LuNi_2B_2C$ single crystals [2]. The characteristic values of the fitting parameters for the measured contacts are presented in Table I both for the one- as well for the two-gap approach.

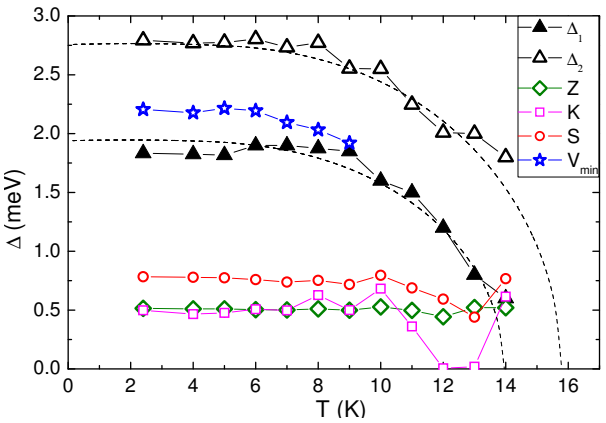

FIG. 2: Temperature dependencies of the fitting parameters: superconducting gaps Δ_1, Δ_2 (triangles), barrier parameter Z (diamonds), contribution (weight factor) K of Δ_1 (squares), scaling parameter S (snowflakes), position of minima in dV/dI (stars) for the PC from Fig. 1. The broadening parameter Γ is equal to zero. The dotted lines represent the BCS-like gap behavior.

TABLE I: Average, minimal and maximal values of the superconducting gap Δ, the "smearing" parameter Γ and the "barrier" parameter Z retrieved from the one- and two-gap approach.

	Δ(meV)	Γ(meV)	Z	$2\Delta/kT_c$
Average	**2.6±0.2**	**0.54±0.2**	**0.48±0.06**	**3.8±0.3**
Minimal	2.12	0.22	0.32	3.21
Maximal	2.85	0.9	0.57	4.16

	Δ_1(meV)	Γ_1(meV)	Δ_2(meV)	Γ_2(meV)	Z
Average	**2.14±0.36**	**0.36**	**3±0.27**	**0.25**	**0.47**
Minimal	1.65	0	2.6	0	0.32
Maximal	2.6	0.74	3.45	0.73	0.54

PCS of quasiparticle excitations

As it was mentioned in the introduction, the PC spectroscopy makes it possible to study the electron-phonon interaction (EPI). The second derivative of the $I(V)$ curve of the ballistic contact at low temperatures is directly proportional to the PC EPI function $\alpha^2F(\omega)$ [1]. The latter can be expressed using measurable signals as

$$\alpha^2F(\omega) = \frac{3}{2\sqrt{2}}\frac{\hbar v_F}{ed}\frac{V_2}{V_1^2}, \qquad (2)$$

where e is the electron charge, d is the PC diameter, v_F is the Fermi velocity, V_1 and V_2 are the rms amplitude of the first and the second harmonics of the modulating signal respectively, which are proportional to the first $\mathrm{d}V/\mathrm{d}I$ and the second $\mathrm{d}^2V/\mathrm{d}I^2$ derivatives of the $I(V)$ curve of the investigated PC, respectively.

In Fig. 3, the d^2V/dI^2 curves of the PC from Fig. 1 are shown. We applied the magnetic field to suppress the superconductivity in the PC, because the magnitude of the superconducting features in the PC spectra near zero bias is much larger than that of the maxima caused by the EPI. As we can see from Fig. 3, the magnetic field of 7 T was not high enough to suppress the superconductivity completely ($B_{c2} \approx 9$ T at 2 K [7]) and the huge feature at bias about 2 mV is due to the residual superconductivity. Besides the feature due to the superconducting gap, a clear-cut maximum slightly above 8 mV and a more smeared one at around 15 mV are well distinguished (see Fig. 3). The mentioned peaks correspond to the low energy phonon maxima in the phonon DOS of $LuNi_2B_2C$ [11] (see Fig. 3, symbols). Whereas the high energy part of the obtained PC spectra contains no visible features. In Fig. 4, we present a set of d^2V/dI^2 curves for different PCs measured up to bias voltages of about 80 mV. By analyzing all of obtained PC spectra (about 30) with the visible phonon features, we have found, that the d^2V/dI^2 curves of the $LuNi_2B_2C$ PCs display phonon maxima at 8.5±0.4 mV and 15.5±1.0 mV (averaged for 8 PC) similarly to the PC from Fig. 3. In contrast they do not contain contributions from the other phonon peaks at 23, 33, and 50 mV observed in the phonon DOS of $LuNi_2B_2C$ (shown in Fig. 4 by the circle symbols). Only the PC with $R = 3.7\,\Omega$ in Fig. 4 shows a weak hump at 23 mV. The reason for the absence of contribution at high-energy phonon maxima could be a deviation from the ballistic (spectroscopic) regime at higher voltages due to increase

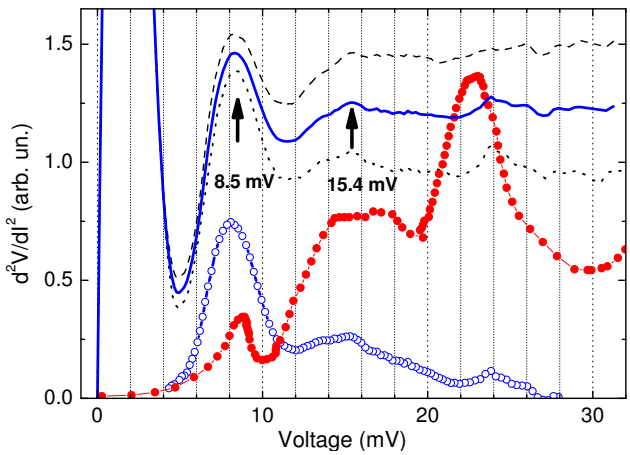

FIG. 3: Averaged for both polarities second harmonic signal $V_2 \propto d^2V/dI^2$ (solid curve) measured in magnetic field of 7 T for the PC from Fig. 1 taken at $T = 2.4$ K in comparison with the phonon DOS for $LuNi_2B_2C$ [11] (solid circles). The open circles show d^2V/dI^2 with subtracted background in the form similar to $\log(V)$ above 6 mV (see also Fig. 4). Dashed and dotted curves show raw data of d^2V/dI^2 for this contact for two bias polarity.

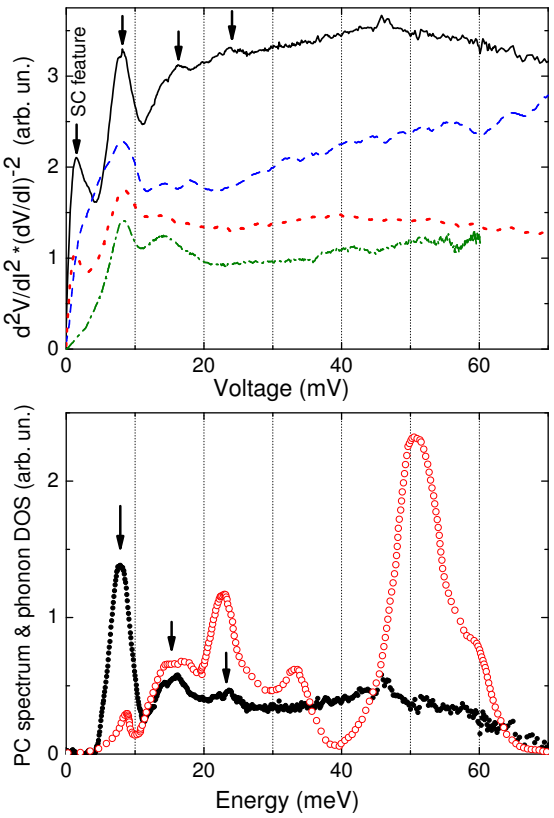

FIG. 4: Upper panel: Reduced second harmonic signal V_2/V_1^2 [$\propto d^2V/dI^2(dV/dI)^{-2} = R^{-1}dR/dV$, where $R = dV/dI$] measured in magnetic field of 9 T for different PCs with R = 3.7, 5.6, 4.7 and 2.9 Ω (from top to bottom) taken at T = 2.2 - 4.2 K. Long-dash line shows tentative background behavior for the upper curve in the form $a\log(bV)+c$. Bottom panel: PC spectrum (solid circles) with subtracted background for the contact with R = 3.7 Ω along with the phonon DOS for $LuNi_2B_2C$ [11] (open circles).

of the EPI and a shortening of the inelastic mean free path of electrons. It is worth to mention that in [12, 13] phonon softening of two branches was observed below T_c. In our case, we did not observe these modes around 4-5 meV, probably because we have measured PC EPI spectra by suppressing the superconducting state, while in other case a huge gap maximum makes it impossible to see any other features below 5-6 meV (see, e.g., Fig. 3, where the superconducting state is not fully suppressed). Another interesting question arises, whether it is possible to separate the contribution of each (two) bands to the PC EPI spectrum? The problem here is similar to the analogous one raised in the tunneling spectroscopy in [14], where the authors concluded that it is not possible to obtain several band splitted EPI functions from a single function of the tunnel current.

After subtraction of the background from the measured PC spectra we established the EPI function according to (2) (using maximal $v_F = 3.6\cdot10^7$ cm/s calculated for $LuNi_2B_2C$ [9]) and estimated the EPI parameter $\lambda = 2\int\alpha^2F(\omega)\omega^{-1}d\omega$. The latter even for the spectra with the maximal intensity was found to be not bigger than 0.1. This value is an order of magnitude smaller than the λ values between 0.5 and 1 from dHvA data for the c - direction of $LuNi_2B_2C$ [16, 17] or $\lambda \approx 0.5-0.8$ estimated from STM measurements on $LuNi_2B_2C$ [18]. Such small values of λ could be due to some simplifications of the PC spectroscopy theory where only the free electron model and a single band Fermi surface are used. Some issues of λ evaluation from PC spectra were discussed in [20]. Most of all, the coupling constants estimated from PC spectra should be considered as lower bonds for the coupling constant λ relevant for superconductivity. Here we also would like to mention that the discussed λ parameter is some kind of transport EPI constant and in general its value is different from the Eliashberg EPI constant (see Table 3.1 in [1]), but the difference by one order of magnitude is of course confusing. The calculation of the PC EPI function may shed light on this issue. It will allow to separate the bands contributions, to estimate the integral intensity of the spectrum and to determine the relative contribution of each phonon branch to the PC EPI spectrum. Sure, it would be a very helpful, but also a sophisticated task and it is beyond the scope of this experimental paper.

Another reason for a reduced intensity of the measured PC spectra might be the elastic scattering, which can be larger in the PC core than in the bulk sample due to less perfect surface properties and stresses at the PC formation. As follows from the PC spectroscopy theory, the magnitude of nonlinearity in PC spectra is proportional to l/d [19] in the diffusive regime $l \ll d$. Fig. 5 shows the intensity of the main peak in the PC spectra, which indeed shows strong scattering in the range of about one order. Since we do not see any trend in the intensity of the main peak vs PC resistance which related to the PC size (diameter), we expect that the spectra with the higher intensity are in (or close to) the ballistic regime. Thus, deviation from the ballistic regime of the current flow in the investigated contacts can not be the reason of small λ values, which were calculated for the PCs with the maximal intensity.

Similar low λ values as we found for $LuNi_2B_2C$ were obtained also from the PC spectra of YNi_2B_2C, $HoNi_2B_2C$ [5, 20, 21] and recently for $TmNi_2B_2C$ (not yet published). There is also general similarity of PC spectra for the mentioned compounds which are characterized by a prevailing first phonon maximum (excluding CEF peaks in $HoNi_2B_2C$ and $TmNi_2B_2C$). As mentioned above, by determining λ from PC spectra one can underestimates the superconducting EPI. In this context, a notable large λ_{PC}=0.85 was reported for one special PC spectra of $DyNi_2B_2C$, while λ_{PC}= 0.25 was found for more typical PC spectra of this compound [22]. However, there by calculating λ_{PC} the low energy intensive

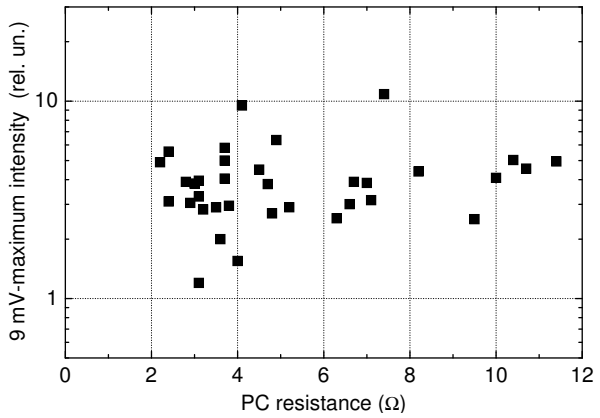

FIG. 5: Scatter in the intensity of the first phonon maximum in d^2V/dI^2 curves for all measured $LuNi_2B_2C$ PCs versus their resistance.

"magnetic" peak at ~ 5 mV was also taken into account, which gives about half of the λ value. Therefore, the concerning EPI contribution to λ in $DyNi_2B_2C$ is again close to 0.1, if we eliminate the mentioned extra high λ value and the "magnetic" peak contribution to λ.

CONCLUSION

We investigated the superconducting energy gap and EPI in $LuNi_2B_2C$ using an epitaxial c-axis oriented film by PC spectroscopy. The mean value of the superconducting gap is found to be about 2.6±0.2 meV ($2\Delta/kT_c = 3.8 \pm 0.3$) in the one-gap model what is very close to the values reported for PC measurements on single crystals [2, 3]. However, the fitting of dV/dI curves favors the two-gap approach which provides strong support for the multiband superconducting state in this compound. For the two-gap approach the averaged gap values are found to be $\Delta_1 \simeq 2.14\pm0.36$ meV and $\Delta_2 \simeq 3\pm0.27$ meV.

For the first time for $LuNi_2B_2C$, we succeeded to measure EPI PC spectra with the distinct phonon peaks at 8.5±0.4 mV and 15.8±0.6 meV. Therefore, we can conclude that these low energy phonons play a preferential role in the pairing mechanism. The EPI spectra are in general similar to those measured for other nickel borocarbides [5, 20–22] showing predominance of the first phonon peak in EPI for all these compounds.

Acknowledgements

Two of the authors (O.E.K., Yu.G.N.) would like to thank the Alexander von Humboldt Foundation for the support, Dr. S.-L. Drechsler for the useful discussions and Dr. K. Nenkov for the technical assistance.

[1] Yu. G. Naidyuk and I. K. Yanson, *Point Contact Spectroscopy*, Springer Series in Solid-State Sciences, Vol.145 (Springer, New York, 2004).
[2] N. L. Bobrov, S. I. Beloborodko, L. V. Tyutrina, I. K. Yanson, D. G. Naugle, and K. D. D. Rathnayaka, Phys. Rev. B **71** 014512 (2005), N. L. Bobrov, S. I. Beloborod'ko, L. V. Tyutrina, V. N. Chernobay, I. K. Yanson, D. G. Naugle, K. D. D. Rathnayaka, Low Temp. Phys., **32**, 489 (2006) [Fiz. Niz. Temp., **32**, 641 (2006)].
[3] Xin Lu, W. K. Park, Jung-Dae Kim, Sunmog Yeo, Sung-Ik Lee, L. H. Greene, Physica B **403** (2008) 1098; Xin Lu, W. K. Park, Ki-Young Choi, Sung-Ik Lee, Sunmog Yeo, Sergey L. Budko, Paul C. Canfield and L. H. Greene, Journal of Physics: Conference Series **150** 052143 (2009).
[4] D. L Bashlakov, Yu. G. Naidyuk, I. K. Yanson, S. C. Wimbush, B. Holzapfel, G. Fuchs and S-L. Drechsler, Supercond. Sci. Technol. **18** 1094 (2005).
[5] D. L. Bashlakov, Yu. G. Naidyuk, I. K. Yanson, G. Behr, S.-L. Drechsler, G. Fuchs, L. Schultz and D. Souptel, J. of Low Temp. Phys., **147**, 336 (2007).
[6] P. Raychaudhuri, G. Sheet,S. Mukhopadhyay and H. Takeya, Physica C, **460-462**, 95 (2007).
[7] T. Niemeier, R. Huhne, A. Kohler, G. Behr, L. Schultz, B. Holzapfel, Journal of Physics: Conference Series **150**, 052185 (2009).
[8] S. V. Shulga, S.-L. Drechsler, G. Fuchs, K.-H. Müller, K. Winzer, M. Heinecke, and K. Krug, Phys. Rev. Lett. **80**, 1730 (1998).
[9] A. K. Bhatnagar, K. D. D. Rathnayaka, D. G. Naugle, and P. C. Canfield, Phys. Rev. B **56** 437 (1997).
[10] G. E. Blonder, M. Tinkham and T. M. Klapwijk, Phys. Rev. B **25**, 4515 (1982).
[11] F. Gompf, W. Reichardt, H. Schober, B. Renker, M. Buchgeister, Phys. Rev. B **55**, 9058 (1997).
[12] P. Dervenagas, M. Bullock, J. Zarestky, P. Canfield, B. K. Cho, B. Harmon, A. I. Goldman, and C. Stassis, Phys. Rev. B. **52**, R9839 (1995).
[13] C. Stassis, M. Bullock, J. Zarestky, P. Canfield, A. I. Goldman, G. Shirane and S. M. Shapiro, Phys. Rev. B. **55**, R8678 (1997).
[14] O. V. Dolgov, R. S. Gonnelli, G. A. Ummarino, A. A. Golubov, S. V. Shulga, and J. Kortus, Phys. Rev. B **68**, 132503 (2003).
[15] S. B. Dugdale, C. Utfeld, I. Wilkinson, J. Laverock, Zs. Major, M. A. Alam and P. C. Canfield, Supercond. Sci. Technol. **22** 014002 (2009).
[16] B. Bergk, V. Petzold, H. Rosner, S.-L. Drechsler, M. Bartkowiak, O. Ignatchik, A. D. Bianchi, I. Sheikin, P. C. Canfield, and J. Wosnitza, Phys. Rev. Lett. **100**, 257004 (2008); B. Bergk and J. Wosnitza, Low Temp. Phys., **35** 686 (2009) [Fiz. Nizk. Temp. **35**, 872 (2009)] .
[17] T. Isshiki, N. Kimura, H. Aoki, T. Terashima, S. Uji, K. Yamauchi, H. Harima, D. Jaiswal-Nagar, S. Ramakrishnan, and A. K. Grover, Phys. Rew. B **78** 134528 (2008).
[18] P. Martinez-Samper, H. Suderow, S. Vieira, J. P. Brison, N. Luchier, P. Lejay, and P. C. Canfield, Phys. Rev. **67** 014526 (2003).
[19] I. O. Kulik and I. K. Yanson, Sov. J. Low Temp. Phys. **4**, 596 (1978) [Fiz. Nizk. Temp. **4**, 1267 (1978)]
[20] Yu. G. Naidyuk, O. E. Kvitnitskaya, I. K. Yanson, G. Fuchs, K. Nenkov, A. Walte, G. Behr, D. Souptel, and

S.-L. Drechsler, Phys. Rev. B **76**, 014520 (2007).
[21] I. K. Yanson, V. V. Fisun, A. G. M. Jansen, P. Wyder, P. C. Canfield, B. K. Cho, C. V. Tomy, and D. McK. Paul, Low Temp. Phys., **23**, 712 (1997) [Fiz. Nizk. Temp., **23**, 951 (1997)].
[22] I. K. Yanson, N. L. Bobrov, C. V. Tomy, D. Mc. K. Paul, Physica C **334**, 152 (2000).